

**PATENT** 

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re:

Patent Application of

Jawahar C. Parekh et al.

Group Art Unit: 1713

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Serial No.:

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Examiner: Peter D.

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HALOGEN SCAVENGERN FOR

No. GC REH 017

## <u>AMENDMENT</u>

This is in response to the Office Action mailed on September 16, 2003 (Paper No. 3) in the above identified application.

## Extension of Time to Respond

Applicants request an extension period of one month, expiring January 16, 2004, thereby extending the period for response set to expire on December 16, 2003.

The Commissioner is authorized to charge the requisite extension fee of \$110.00 for the one-month extension to account no. 070825 for which a duplicate copy of this page and the signatory page is attached.

## The Rejection of the Claims

The Examiner has rejected claims 1-20, all the claims in the application under 35 U.S.C. § 103(a) as being unpatenable over Miyata et. al., U.S. Patent No. 4,284,762 or Miyata, U.S. Patent No. 4,675,356 on the grounds that these cited patents disclose methods of inhibiting corrosion of olefin polymers and render the present invention obvious because these patents show the utilization of hydrotalcite compounds which are formulated from elements as well as molar ratios that would be obvious to those claimed by the applicant. In the rejection Examiner alludes to specific portions of the '762 and

'356 Patents and concludes that this patent provides one of ordinary skill in the art with sufficient direction so as to formulate the invention claimed.

The rejection of the claims is respectfully traversed and, for the reason set forth hereinbelow, the Examiner is respectfully requested to reconsider the applied grounds of rejection because of distinctions, i.e., the non-obvious differences, in the claimed invention over the disclosure and teaching of the two cited Miyata '762 and '365 Patents.

With regard to U.S. Patent No. 4,284,762 (the '762 patent), there are many differences between the claimed solid solutions and the compounds described in the '762 patent. As an example the Examiner's attention is respectfully directed to the fact that the '762 patent claims are limited to the use of hydrotalcites having the formula

$$Mg_{(1-x)}Al_x(OH)_2Ax/n^{n-}$$
  $\bullet mH_2O$ 

wherein  $0 < x \le 0.5$ ; m is a positive number, and  $A^{n-}$  represents an anion having a valance of n. Such hydrotalcites have a BET specific surface area of not more than 30 m<sup>2</sup>/gm and an average secondary particle size of not more than 3 microns. Also, it is clear to one skilled in the art by reference to the above formula that where A is  $CO_3^{=}$ , the Al/CO<sub>3</sub> ratio is always 2:1. This is substantiated by the examples contained in Column 10, Table I of the '762 patent. However, the Al/CO<sub>3</sub> ratio of the claimed invention varies from 0.67 to 1.67 and does not include 2, a fact, the significance of which is not obvious from the prior art.

Reference to the '762 patent (see column 4, lines 25 to 34) discloses as a requirement that the hydrotalcite have a BET surface area of not more than 30 m<sup>2</sup>/gm, preferably not more than 20 m<sup>2</sup>/gm, and, in particular, not more than 15 m<sup>2</sup>/gm. Also required by that disclosure is an average particle size of not more than 5 microns, and

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optimally not more than 1.5 microns and preferably a crystallite size in the <003>

direction determined by an x-ray diffraction method of at least 600Å, preferably at least

1000Å.

These distinctions are supported by the examples 1 through 5 in Column 10,

Table I of the '762 patent. Crystallite size in the <003> direction is from 620Å to 1650Å,

average secondary particle size is 0.5 to 1.4 microns and BET surface area is 7-20

 $m^2/gm$ . The same table shows that when crystallite size in <003> direction is in the range

of 420Å to 650Å, particle size in the range of 3.2 to 7 microns and a BET surface area is

25 - 35 m<sup>2</sup>/gm, corrosion resistance is less and yellowing resistance is also less (pale

yellow vs. white). These improvements are not obvious from the patent disclosure. It is

thus seen that a hydrotalcite product meeting the said formula should have BET surface

area of 15 m<sup>2</sup>/gm, particle size of 1.5 microns, and crystallite size in <003> direction of

1000Å.

The product of the present invention, on the other hand, is micronized to reduce

particle size below five microns which is required because of its use in the manufacture

of films where thickness can be 5 microns or less. It is not necessary to reduce the

particle size of the compound below 10 microns to achieve the desired corrosion

inhibition and/or color control.

The process parameters optimized for the manufacture of the product of this

invention yield a product with a BET surface area in the range of 25 - 35 m<sup>2</sup>/gm.

Pursuant to the invention, the average BET value of the product made in the lab, in the

pilot plant, and in production, is 25.2 m<sup>2</sup>/gm, a result which is approximately double that

of Miyata's preferred value. See example CEx2 in Column 10, Table I of the '762 patent

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which shows that a BET surface area of 25 m<sup>2</sup>/gm, and a result in which the corrosion

resistance and color are inferior. Another important requirement of the product of the

'762 disclosure is that the crystallite size in the <003> direction as determined by an x-

ray diffraction method should be at least 600Å and preferably at least 1000Å.

The Miyata, et. al. '762 patent (See column 4, lines 35-45) also states that

hydrotalcites generally available on the market have a crystallite size of as small as about

100Å to about 300Å, a large crystal strain, a strong aggregating property (an average

secondary particle size of at least about 10 microns, usually about 20 to about 70

microns), and a crystallite size in the <003> direction determined by an x-ray diffraction

method of not more than 300Å. The corrosion resistance and coloration inhibition cannot

be achieved with such hydrotalcites.

Example 9 and comparative example 1 of the current patent application has

demonstrated that the novel product of this invention with Al/CO<sub>3</sub> ratio of 1.39:1, having

a crystallite size in the <003> direction of 190Å, BET surface area of 30.5 m<sup>2</sup>/gm is a

superior halogen scavenger and more consistent in color inhibition than that of the

Miyata '356 Patent.

From the foregoing, it is apparent that the product of the present invention is not

obvious because, as the application teaches a crystallite size in <003> direction is not

essential as taught by the patents relied upon by the Examiner. In other words, despite

the chemical, structural and physical differences, the novel product of the present

invention is at least equal and more likely superior to that of they Miyata teaching.

Complete X-Ray diffraction analysis shows the significant differences in

interplaner spacings which are believed to be due to structural differences between the

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product of this invention and that disclosed as the prior art product as evidenced by

reference to attachment I. With reference to this attachment, in the Examiner's discretion

applicants are prepared to re-submit the attachments referred to herein, accompanied by a

declaration.

Further evidence of the differences between the claimed solid solutions and the

compounds of the '762 patent can be seen by infrared spectrum (see attachment II) of the

novel compound of the present invention that displays a split absorption band in the range

of 1415-1480 cm<sup>-1</sup>, showing a bidentate coordination for carbonate ion. This frequency

split is absent in the hydrotalcite of the reference patents disclosure.

Additionally, the presence of a frequency split at 440-390 cm<sup>-1</sup>, which is

probably due to Al-O and Mg-O vibration of the octahedral sheets in the hydrotalcite

product obtained in accordance with the '356 disclosure indicates major structural

differences. [Ref. G. Brown and M. Gastuche, Clay Miner., 7, 193 (1967)] between the

product of this invention and that of the hydrotalcite disclosed by Miyata.

Differential thermal analysis of the product of this invention and the hydrotalcite

of '762 patent shows significant differences (see attachment III). These differences are

due to different binding forces between anion and the basic layer.

The structural differences are also observed by the <sup>27</sup>Al NMR between the product

of this invention and Kyowa's hydrotalcite (DHT 4A®). The <sup>27</sup>Al NMR spectra of DHT

4A® show a simple relatively narrow peak at 8.6 ppm corresponding to octahedral

aluminum (AlO<sub>6</sub>) (see attachment IV). The spectra of the present invention shows two

types of peaks: a relatively narrow signal at 8.4 ppm and a much broader signal at 3.1

ppm. From the deconvolutions, the relative amounts can be computed. The ratio of the

8.4 to 3.1 ppm signals is 1:1.418 and confirms the presence of different aluminum species and further supports the fact that the chemical structure of the product of the present invention is different.

An important advantage of the present invention resides in the fact that prior art hydrotalcite materials which are used for corrosion inhibition and color control of polyolefins are produced by relatively more severe hydrothermal treatment such as heating at a temperature of 150°C to 250°C for a period of 5 to 30 hours ('762 patent). Whereas those of the process of the present invention are more economical because such do not require the severe hydrothermal treatment condition employed by the referenced patents.

Referring in particular to the disclosure of U.S. Patent No. 4,675,356 (the '356 patent), it is seen that there are many differences between the solid solutions disclosed therein and those claimed by the present invention.

The preparation of hydrotalcite-like compounds (i.e., synthetic hydrotalcites) requires the choice of optimal composition and preparation methods, such as precipitation conditions, types of reagents, aging, washing, hydrothermal treatments, drying, calcination, etc., in order to make a product of choice with different structures. Depending upon the synthesis and processing technique employed, different solid solutions will be formed. The term "solid solution" as used in the present application, refers to a homogeneous crystalline phase composed of several distinct chemical species, occupying the lattice points at random and existing in a range of concentrations. Thus, as shown by the formulas and methods in the present claims, it is important to note that the solid solutions of the present invention consist essentially of aluminum species and

magnesium species, and they exclude the presence of additional bivalent or trivalent metals except only for trace metal impurities.

It is evident, accordingly, that products of the Miyata '356 patent, represented by the formula:

$$(M_1^{2+})_{v1}[(M_2^{2+})_{v2}]_{1-x}M_x^{3+}(OH)_2A_{x/n}^{n-} M_2O$$

wherein  $M_1^{2+}$  represents at least one divalent metal cation selected from the group consisting of Mg, Ca, Sr and Ba;  $M_2^{2+}$  represents at least one divalent metal cation selected from the group consisting Zn, Cd, Pb and Sn;  $M^{3+}$  represents a trivalent metal cation (e.g.,  $A1^{3+}$ );  $A^{n-}$  represents an anion having a balance of n; and x, yl, y2 and m are positive numbers represented by the following expressions:  $0 < x \le 0.5$ , 0 < y1, 0 < y2,  $0.5 \le y1 + y2 < 1$ , and  $0 \le m \le 2$ , would have totally different chemical structures than the products of this invention.

Attention is directed to Miyata '356 at column 7, line 44, wherein it is stated that:

"it is preferred to select hydrotalcite solid solutions subjected to a treatment of removing water of crystallization in which m is zero or nearly 0. The treatment of removing water of crystallization can be easily carried out, for example by treating the hydrotalcite solid solutions at a temperature of about 110°C to about 400°C for about 1 to 40 hours in an atmosphere such as  $N_2$ , or He."

The present invention imposes no such requirement. Additionally, the water of crystallization is about 6 moles (actual values are 6.69, 7.86, 5.76, 5.0, 4.9 for Examples 1, 4, 6, 7, 8, respectively) for the hydrotalcite of this invention (see attachment V). Also, by reference to column 7, line 26 of Miyata '356 it is seen that the crystal grain size and distribution are different from those of the present invention.

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It is respectfully submitted that it appears that the Examiner has overlooked the

fact that the Miyata '356 patent requires at least two dissimilar divalent metal cations,

namely M<sub>1</sub><sup>2+</sup>, which may be selected from Mg, Ca, Sr and Ba, and M<sub>2</sub><sup>2+</sup>, which may be

selected from Zn, Cd, Pb and Sn. In contrast, the hydrotalcite solid solution of the

present invention requires the single divalent metal cation magnesium.

In this connection it is to be noted, accordingly, that the presence of two

dissimilar divalent metals (one selected from periodic table Group II and another selected

from periodic table Group IIb and IVb) is critical for the compositions of the '356 patent

(see column 4, line 52-68 and column 5, lines 11-25). This is a limitation and a clear

distinction that is not applicable to the present invention.

In the '356 patent the presence of the two dissimilar divalent metals is necessary

to inhibit the coloration or degradation and provide the heat resistance or weatherability

of synthetic resins (see column 4, lines 57-61). In the present invention, however, the

single divalent metal (Mg) and a high carbonate level provide the hydrotalcite solid

solutions capable of mitigating the corrosion properties of the polymers tested (see for

example the data presented in Tables E and F of the present application) and give

consistent color throughout five extrusion passes (see Table B of applicants'

specification).

The solid solutions of the '356 patent are defined by the structural formula at

column 6, line 23, and explained at column 6, line 24 to column 7, line 2. While there are

a large number of variables in the structural formula, all of the disclosed compounds have

an Al/CO<sub>3</sub> molar ratio of 2. Thus, the number of carbonates is always half the number of

aluminum ions, so that the mole ratio of Al/CO3 is fixed at 2. This contrasts with the

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solid solutions of the present invention, wherein the Al/CO<sub>3</sub> ratio varies from 0.67 to 1.67, and does not include two (2) as does the system of the '356 patent. Furthermore, there is no disclosure, teaching or suggestion in the '356 patent as to how to vary the Al/CO<sub>3</sub> ratio in a solid solution. Thus, even if one of ordinary skill in the art were somehow motivated by the '356 patent to vary the Al/CO<sub>3</sub> ratio, the '356 patent is completely silent on how to achieve such variation. In contrast, the claimed preparative methods of Applicants' invention clearly disclose and teach the starting material stoichiometry to arrive at the solid solutions being claimed and which have Al/CO<sub>3</sub> ratios outside that of the '356 patent.

It is further noted that one of ordinary skill in the art would not read the '356 patent as a teaching or suggestion to prepare solid solutions according to Applicants' claimed invention, which has a completely different structural formula from that set forth in the '356 patent. Applicants submit, accordingly, that the disclosure in the '356 patent does not teach, nor make obvious, Applicants' claimed invention, nor does that disclosure contain any reasonable suggestion to motivate one skilled in the art to arrive at Applicants' claimed invention. A reconsideration and withdrawal of the rejections in view the cited prior art therefore are respectfully requested.

In view of the foregoing enumerated distinctions, Applicants respectfully submit that the invention claimed in the above-identified application is neither disclosed nor rendered obvious by either or both of the two Miyata U.S. Patent Nos. 4,284,762 or 4,675,356, nor from any of the prior art of record singly or in combination.

Applicants inventive contribution comprises the ability to produce excellent polymer characteristics by way of a distinctly different hydrotalcite solid solution

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composition. For this reason it is submitted that the Examiner's observation that the

intended function of the hydrotalcite compositions is the same and that one of ordinary

skill in the art would have a reasonable expectation of success when utilizing hydrotalcite

compounds having molar ratios as those presently is submitted as tenuous; applicants are

not claiming a method of using the hydrotalcite compounds, but are claiming new

hydrotalcite compositions per se. This specifically is a non-obvious advance that

contributes materially to the art.

Applicants have in fact demonstrated unexpected results in that they provide a

hydrotalcite solid solution composition which omits the requirement of two dissimilar

divalent metals and significantly raises the carbonate ion concentration, as compared to

those of the '356 patent while still deriving excellent, and in fact improved, mitigation of

corrosion and consistency of color in treating various polymer compositions. (See for

example, the Performance and Comparative Examples at pages 22-29 of Applicants'

specification).

In view of the above amendments and remarks, it is submitted that the claims in

the application fully comply with the requirements of 35 U.S.C. §103(a), in that the

claims are not obvious from the prior art and consequently do distinguish over the prior

art of record.

A reconsideration and withdrawal of the rejections and an early notice of

allowance are respectfully solicited.

Respectfully submitted,

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